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6. AUTHOR(S)

S.R. HARTMANN

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)

COLUMBIA UNIVERSITY
PHYSICS DEPARTMENT
NEW YORK, N.Y. 10027

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We have studied the time evolution of the radiation field spontaneously emitted by an ensemble of many atoms as they proceed from some particular excited atomic state to a lower one (initially unpopulated) and then on to the ground state. A short laser pulse, two-photon resonant with an even parity optical transition, produces the excited atomic population which is forbidden (by symmetry) to radiate directly back to the ground state and must therefor radiate first to an intermediate state. If the gain is high, laser action should rapidly

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deplete the excited state and with sufficient population build-up on the intermediate state one can expect laser action to appear on the transition to the ground state. This scenario becomes more interesting when the "short" laser pulse coherently excites the atoms into a coherent superposition of the ground and uppermost resonant state. Now laser action on the uppermost transition directly induces laser action on the lower transition even though no population inversion is present there. The effect is to modify the laser action on the uppermost transition. We have used a mode locked laser to produce pulses short enough to coherently excite the $6D_{3/2}$-$6S_{1/2}$ transition in Cs vapor. Our observations indicate that the induced coherence inhibits superfluorescence and that laser action on the uppermost transition in the backwards direction (opposite to the direction of the short excitation pulse) strongly perturbs this interference. The theoretical analysis is highly nonlinear and numerical techniques are applied.

TWO-PHOTON COOPERATIVE CASCADE SUPERFLUORESCENCE

FINAL REPORT

Prepared by:
S. R. HARTMANN

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Submitted by:
Columbia University
Physics Department
New York, NY 10027
(212) 854 3272

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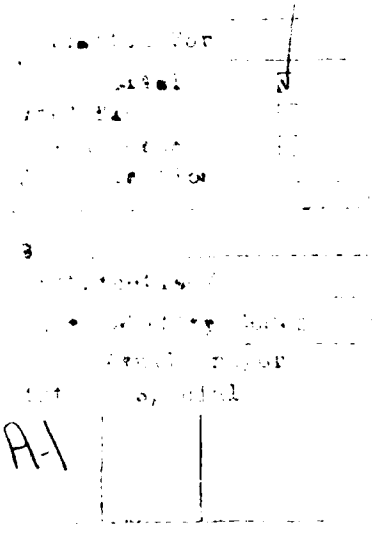

ABSTRACT

We have studied the time evolution of the radiation field spontaneously emitted by an ensemble of many atoms as they proceed from some particular excited atomic state to a lower one (initially unpopulated) and then on to the ground state. A short laser pulse, two-photon resonant with an even parity optical transition, produces the excited atomic population which is forbidden (by symmetry) to radiate directly back to the ground state and must therefor radiate first to an intermediate state. If the gain is high, laser action should rapidly deplete the excited state and with sufficient population build-up on the intermediate state one can expect laser action to appear on the transition to the ground state. This scenario becomes more interesting when the "short" laser pulse coherently excites the atoms into a coherent superposition of the ground and uppermost resonant state. Now laser action on the uppermost transition directly induces laser action on the lower transition even though no population inversion is present there. The effect is to modify the laser action on the uppermost transition. We have used a mode locked laser to produce pulses short enough to coherently excite the $6D_{3/2}$-$6S_{1/2}$ transition in Cs vapor. Our observations indicate that the induced coherence inhibits superfluorescence and that laser action on the uppermost transition in the backwards direction (opposite to the direction of the short excitation pulse) strongly perturbs this interference. The theoretical analysis is highly nonlinear and numerical techniques are applied.

STATEMENT OF PROBLEM STUDIED

The problem is to investigate the cascade superfluorescence that develops when an ensemble of three-level atoms is initially excited by a two-photon excitation resonant with the ground and uppermost state [1]. The interest is for short pulse excitation which makes the excitation coherent. Because the intermediate state is initially unpopulated superfluorescence develops on the upper transition [2]. When the excitation is coherent superfluorescence is also expected to develop by coherence transfer on the lower transition even without population inversion. The action of the superradiant emission on the lower transition modifies the superradiant development on the upper transition and provides an interesting study for understanding the interaction of light and matter.

DISCUSSION OF EXPERIMENTS

We have excited the $6D_{3/2}$-$6S_{1/2}$ transition in Cs vapor with a short, 5 ps, resonant (two-photon) excitation pulse and have observed coherently excited two-photon cascade superfluorescence on the $6D_{3/2}$-$6P_{1/2}$ and $6P_{1/2}$-$6S_{1/2}$ transitions. We have excited the $6P_{1/2}$-$6S_{1/2}$ transition with a similarly short (single-photon) excitation pulse and have observed photon echoes [3] on the same transition. We have also measured fluorescence on the $6D_{3/2}$-$6P_{1/2}$ transition (after (two-photon) pulse excitation on the $6D_{3/2}$-$6S_{1/2}$ transition) at sufficiently low densities so that superfluorescence does not occur. In this latter experiment we find that when measured against the energy of the excitation pulse the intensity of the fluorescence is too small by a factor of ten whereas the related two-photon pulse power necessary to excite it is too large by a factor of ten. This is interesting in as much as the cooperative cascade emission experiments of Okada, Ikeda, and Matsuoka [1] found a similar discrepancy but a factor of ten larger still. We have compared the excitation pulse autocorrelation trace with the pulse spectrum and find that the latter implies a pulse width a factor of two larger. The analysis of the autocorrelation response assumes the pulse is transform limited; the discrepancy shows that it isn't. We believe there may be chirping effects. Their manifestation on the two-photon transition is somewhat complicated and suggests that it might be better to look at the corresponding fluorescence experiment on the single photon $6P_{1/2}$-$6S_{1/2}$ transition.

The cascade superfluorescence experiments haven't been yet shown the reproducibility which we require for a thorough analysis. The elements of the inhibition we are looking for are however evident in our data. An example is to be found in the run at 200 °K which we show in Figures 1 and 2. The data of Fig. 1 shows that at low excitation energy we obtain laser action in the backward direction and none in the forward direction. The superfluorescence is

clearly inhibited. This behavior is to be contrasted with the action on the lower transition at 894 nm shown in Fig. 2. In this case we note that we obtain a large amount of radiation in the forward direction and only a small amount in the backward direction. This behavior is consistent with the behavior observed on the upper transition. It is precisely because there is enhanced radiation in the forward direction on the lower transition that the radiation in the forward direction on the upper transition is inhibited. The forward radiation on the lower transition robs the coherent moment phased to radiate in the forward direction at the upper transition. No such robbing occurs on the associated coherent moment for the backward moment. Thus spontaneous emission from the $6D_{3/2}$ level builds up in the backward direction at a much higher rate than in the forward direction. At large excitation energies there is a reversed behavior. Now the energy in the forward direction exceeds that in the backward direction. This is an unexpected result and we are not clear about its origin, probably some kind of parametric amplification effect.

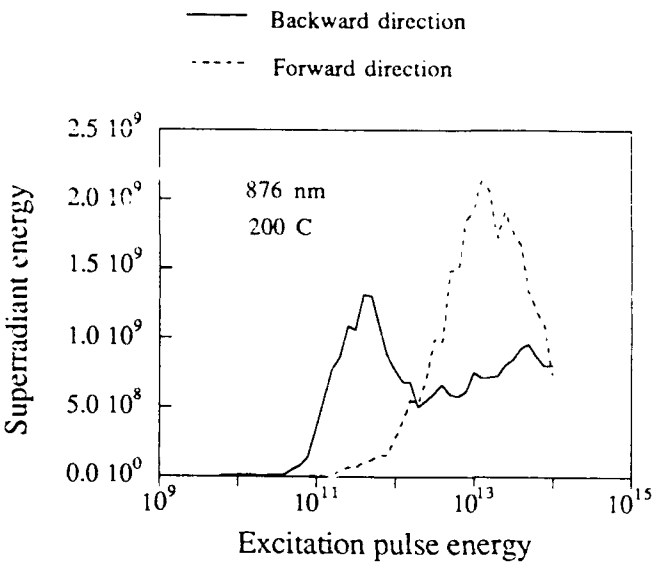

Fig. 1 Superfluorescence output energy (photons / pulse) at the upper optical transition at 876 nm is plotted as a function of the energy of the 5 ps two-photon excitation pulse. The sample temperature is 200 °C, the number density is 2×10^{15} cm^{-3}, αL=7,000 on the 876 nm transition.

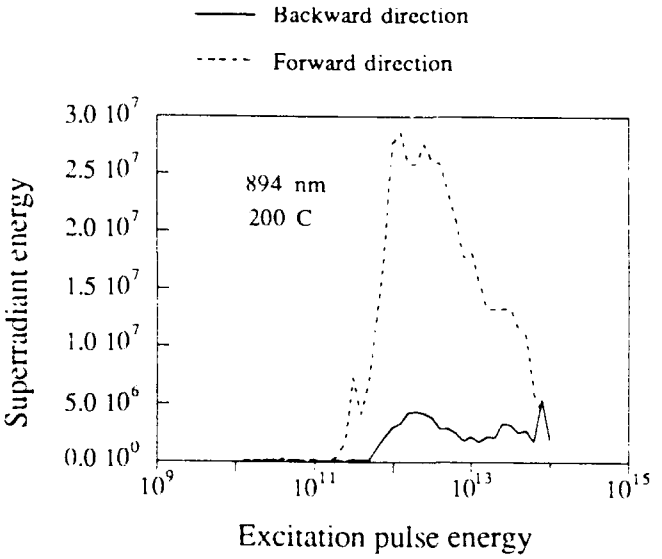

Fig. 2 Superfluorescence output energy (photons / pulse) at the lower optical transition at 894 nm is plotted as a function of the energy of the 10 ps two-photon excitation pulse. The sample temperature is 200 °C, the number density is 2×10^{15} cm^{-12}.

The energy units in Figs. 1 and 2 are in terms of

the number of photons per excitation pulse. It is of interest to know how the excitation energy corresponds to pulse area. We calculate that for our sample geometry approximately 10^{12} photons corresponds to a π pulse. Our fluorescence experiments show that it is more correct to associate 10^{13} photons with a π pulse. This anomaly was noted earlier and appears in other work also. When the log of the fluorescence intensity is plotted as a function of the pump energy there is a distinct kink in the plot at the pump energy corresponding to a π pulse. The excitation beam has a gaussian transverse spatial profile and the Rabi oscillations are effectively averaged out. Increasing excitation energy always yields increased fluorescence however when the excitation pulse area on axis reaches π the rate of increase is diminished markedly and leads to the appearance of a kink. The location of the kink is easily found and we used it to determine the effective pulse area.

The coherent nature of the two-photon excitation pulse is essential to a proper interpretation of our experiment. For that reason we have performed photon echo experiments on the lower optical transition at 894 nm. To our surprise we found that we obtained large fluctuations in photon echo intensity even though the excitation energy was stable. This is the sort of observation we had made in our superfluorescence experiments but there it was expected. After all, in superfluorescence it is the spontaneous emission which triggers the resulting coherent radiation. This by its very nature leads to large fluctuations [4]. In the photon echo case spontaneous radiation plays no role and we expect the signals to be stable. Further experimentation showed that if we monitored the energy of the excitation pulse by filtering it through a grating spectrometer (bandwidth .25 Å) then the echoes measured against this energy rather than the magnitude of the response of a broadband diode detector became dramatically more stable. This is further evidence that the excitation pulses are not transform limited. We still have a large residual fluctuation of about 50 % which we attribute to instabilities in the phase characteristics of the excitation pulse.

The difficulties associated with the theoretical analysis of the cascade superfluorescence problem are formidable. Ikeda, Okada, and Matsuoka [5,6] treat the problem extensively but they do so only in the linear regime and even there they do not obtain analytic expressions for the time development. It seems that the only viable way to treat this problem is numerically and we have begun to do so. (In this aspect of the work we are collaborating with a colleague Professor J. Manassah with whom I have published extensively on other problems.) The linear regime assumes that one can neglect temporal variation in the diagonal density matrix elements. To obtain some feeling for the validity of this approach we have considered the problem in which a short pulse of area θ_0 at the frequency, $\Omega_c - \Omega_b$, of the upper transition is applied to the input face of a sample of fixed size and concentration. We calculate the

amplification and reshaping of this input pulse. The sample consists of three-level atoms with states $|a\rangle$, $|b\rangle, |c\rangle$, and energies $\hbar\Omega_a < \hbar\Omega_b < \hbar\Omega_c$. We assume the sample has been prepared so that the initial density matrix elements are ρ_{aa} and $\rho_{cc} = 1 - \rho_{aa}$ with either $\rho_{ac} = 0$ (incoherent case) or $\rho_{ac} = \sqrt{\rho_{cc}\,\rho_{aa}}$ (coherent case). The problem is akin to cascade superfluorescence where a zeropoint noise pulse triggers the superradiant output. The gain on the upper transition increases linearly with ρ_{cc} and for our calculation it is $\alpha L = 200\,\rho_{cc}$. In Fig. 3 we show the amplified pulse area in both the incoherent and coherent cases. The incoherent case follows $\theta = \theta_0 e^{\alpha L/2}$ until θ becomes of the order of π at which point the linear approximation breaks down. The coherent case is somewhat inhibited but not to the extent we might have expected from the analysis of Ikeda *et al* [5]. Interestingly enough the two results converge once the linear approximation breaks

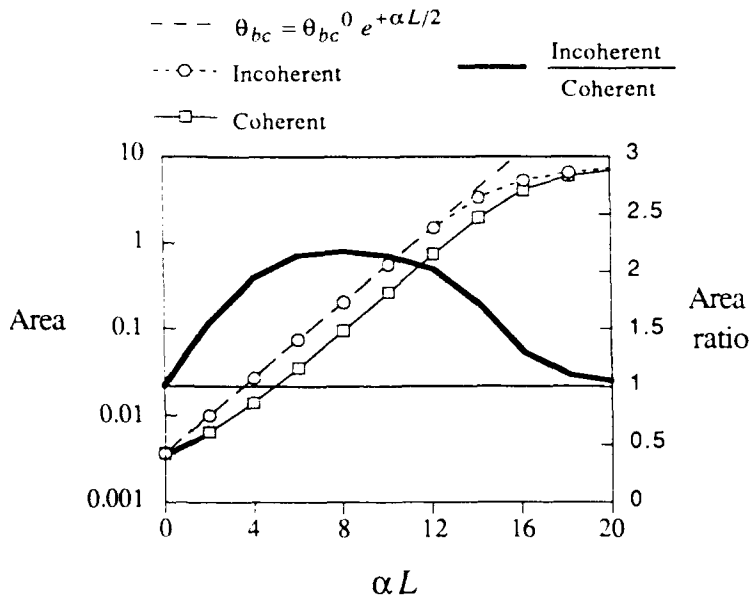

Fig. 3 The amplified pulse (propagating on the upper transition) area at the output face is plotted as a function of the gain for incoherent excitation (open circles) and coherent excitation (open squares). The ratio of the areas (incohernt/coherent) is also shown.

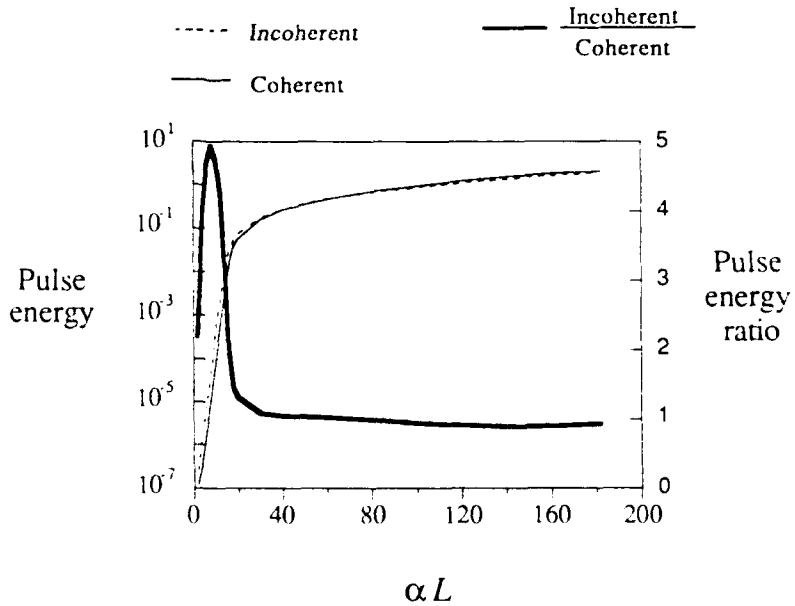

Fig. 4 The amplified pulse (propagating on the upper transition) energy at the output face is plotted as a function of the gain for incoherent excitation (dashed curve) and coherent excitation (solid curve). The ratio of the pulse energies is also shown.

down.

Beyond the linear approximation the initial pulse undergoes severe reshaping and it is not meaningful to follow the pulse area. Instead we follow the amplified pulse energy *cf* Fig 4. Here again we note that once the linear region is passed the coherently excited case can result in just as large an amplification as the incoherently excited one.

The calculations were not meant to mimic the cascade superfluorescence case as the input pulse area we chose was much too large. They are useful as they indicate what one might expect if one applied a trigger pulse on the upper transition. It should be noted that the equalization of output energies outside the linear approximation does not necessarily mean that cascade superfluorescence should then be strong. An examination of the reshaped amplified pulses in this regime shows that they undergo an additional temporal delay in the coherent case. But if they are delayed then they will be unable to compete with the backward directed superfluorescence as it always corresponds to the incoherently excited case and thus precedes the cascade superfluorescence. Of course the major build up of the respective superradiant emissions is at the sample faces at which they exit. This fact may effectively decouple the two modes and allow the cascade signals to develop fully.

SUMMARY OF IMPORTANT RESULTS

1 We observe inhibition of cascade superfluorescence at low excitation energies.

2 Photon echo stability experiments reveal sensitivity to fine structure of excitation pulses.

3 Theoretical calculations indicate that the effect of the coherent two-photon excitation is to produce a delay in the superfluorescence development rather than an inhibition.

PERSONNEL

Dr. David Debeer	Post doc
Hayden Brownell	Grad. student
Xingming Lu	Grad student

PUBLICATIONS AND TECHNICAL REPORTS

There are no publications yet. A theoretical paper is in preparation.

REPORT OF INVENTIONS

None.

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